

Micro-scale CFD study about the influence of operative parameters on physical mass transfer within structured packing elements



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ARTICLE INFO

Article history:

Received 7 February 2014

Received in revised form 12 May 2014

Accepted 25 June 2014

Available online 15 July 2014

Keywords:

CFD

Structured packing

Carbon capture

VOF

Physical mass transfer

ABSTRACT

In this work a VOF-based 3D numerical model is developed to study the influence of several operative parameters on the gas absorption into falling liquid films. The parameters studied are liquid phase viscosity, gas phase pressure and inlet configuration, liquid–solid contact angle and plate texture. This study aims to optimize the post-combustion CO₂ capture process within structured packed columns. Liquid phase viscosity is modified via MEA (i.e. monoethanolamine) concentration. The results show that an increase in liquid viscosity reduces the diffusivity of oxygen within the liquid film thus hindering the efficiency of the process. Higher pressure carries an absorption improvement that can be attractive to be applied in industry. The simulations show that enhanced oxygen absorption rates can be achieved depending on the velocity of the gas phase and the flow configuration (i.e. co- and counter-current). Also, the importance of wetting liquid–solid contact angles (i.e. less than 90°) is highlighted. Poor liquid–solid adhesion has similar effects as surface tension in terms of diminishing the spreading of the liquid phase over the metallic plate. Finally the influence of a certain geometrical pattern in the metallic surface is also assessed.

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1. Introduction

Different techniques exist in order to reduce CO₂ emissions to the atmosphere: renewable energies, efficiency improvement of current energy generation systems and carbon capture and storage (CCS). Renewable energies would be the definitive option to avoid emissions but their current cost is not competitive compared with fossil fuel technologies (Khalilpour, 2013). Recent studies suggest that fossil fuels are expected to play a dominant role during the next few decades for power production (Odenberger and Johnsson, 2010). CCS is the technology that deals with the process of capturing CO₂ from a flue gas mainstream, transport it and store it into geological formations. CCS is thus presented as a valuable option for greenhouse gases (GHGs) reduction in the short-term. Three different approaches exist to mitigate CO₂ emissions: pre-combustion, oxy-fuel technology and post-combustion carbon capture. Pre-combustion extracts carbon from the fuel before combustion and is

based on a gasification process using oxygen followed by a water-gas shift reaction. In oxy-fuel technology the combustion takes place in the presence of pure oxygen giving steam and carbon dioxide as products. The latter can thus be removed by condensation. Post-combustion technologies deal with capturing CO₂ after the combustion of the fossil fuel. The main post-combustion processes are: adsorption, cryogenic separation, membrane absorption and physical and chemical absorption (Wang et al., 2011). Adsorption refers to the physical attachment of the CO₂ molecules to the surface of a solid material such as activated carbon, alumina, zeolites and metal organic frameworks (MOFs). Cryogenic separation works on the basis of condensation. At atmospheric pressure, the dew point of CO₂ lies below −50 °C. This process is commonly used to capture CO₂ when oxy-fuel technologies are applied. Absorption by means of physical membranes has the advantage of presenting compact devices. It is suitable for flue gas streams with high CO₂ concentrations as those encountered in integrated gasification combined cycle (IGCC) power plants and oxy-fuel processes. Physical absorption refers to the dissolution of the gas in liquid solutions such as dimethyl ethers of polyethylene glycols and methanol. A basic requirement is a high CO₂ partial pressure, which does not occur in natural gas power plants. Chemical absorption of acidic gases by means of amine solutions is a common

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Nomenclature

Latin symbols

a	area per unit volume (m^{-1})
C	total concentration (kg m^{-3})
D	mass diffusivity ($\text{m}^2 \text{s}^{-1}$)
f	additional force term in momentum equations ($\text{kg m}^{-2} \text{s}^{-2}$)
F	volume fraction
g	acceleration of gravity (m s^{-2})
k	mass transfer coefficient (m s^{-1})
L	distance traveled by liquid film (m)
M	molecular weight of solvent (kg kmol^{-1})
P	pressure (Pa)
S	mass source term ($\text{kg s}^{-1} \text{m}^{-3}$)
t	time (s)
T	temperature (K)
v	velocity (m s^{-1})
V	molar volume ($\text{m}^3 \text{mol}^{-1}$)
y	mass fraction
z	coordinate along film thickness (m)

Greek symbols

β	plate inclination angle
δ	liquid film thickness (m)
κ	surface curvature (m^{-1})
μ	dynamic viscosity (Pa s)
ρ	density (kg m^{-3})
σ	surface tension coefficient (N m^{-1})
τ	exposure time (s)
ϕ	association parameter of solvent

Subscripts

e	effective
g	gas phase
inj	injection
l	liquid phase

Other symbols

He	Henry's law constant
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process in industry (Littel et al., 1992). It is based on a chemical reaction between CO_2 and a solvent. The weak chemical bonds linking both substances are broken by applying heat in another device called stripper. The chemical reaction allows an increase in the amount of CO_2 removed from the gas stream compared to physical absorption via the enhancement factor. The most common active substances are aqueous solutions of MEA (monoethanolamine, $\text{C}_2\text{H}_7\text{NO}$), MDEA (methyldiethanolamine, $\text{C}_5\text{H}_{13}\text{NO}_2$) and TEA (triethanolamine, $\text{C}_6\text{H}_{15}\text{NO}_3$). The absorption takes place in a device called absorber where the CO_2 -lean solvent is injected at the top section whereas the gas phase enters the reactor through the bottom. Counter-current flow is thus the most common configuration for gas–liquid flow in these devices. The absorber is filled either with a structured packing material, which is formed by corrugated metal sheets, or with random packing. After the chemical reaction, the output is a CO_2 -rich solvent and a flue gas stream that can be emitted to the atmosphere. The CO_2 -rich solvent enters the stripper, where hot steam is applied to separate again the carbon dioxide from the active solution. The amine solution is then recirculated to the absorber whereas the CO_2 stream is ready for the next stages of the process (i.e. transport and storage).

The present study deals with the numerical modeling of the post-combustion carbon capture in a structured packed absorption

column. CFD has been proven to be a suitable tool to reduce the cost associated with the experimental study of multiphase flow phenomena. The limited space between metal sheets makes it impossible to insert any measuring device without interfering with the flow within, which makes numerical simulations attractive in this field. However, the computational capacity available nowadays is limited and the study of post-combustion absorbers is divided in three different scales depending on the aspect to be evaluated: micro-scale, meso-scale and macro-scale (Raynal et al., 2009; Raynal and Royon-Lebeaud, 2007; Sun et al., 2013).

In micro-scale simulations, the gas–liquid flow between metal sheets is tracked using the volume of fluid (VOF) method. Reactive mass transfer can also be implemented and studied with this approach. Meso-scale single-phase simulations use a small set of representative elementary units or REU (i.e. a repeating geometrical pattern formed by the metal sheets) to study the gas dry pressure drop characteristics. According to Larachi et al. (2003) and Said et al. (2011) the pressure drop per unit length for a small number of REUs is the same as for the entire column, which simplifies its computational modeling. Correction factors are applied to obtain the pressure drop of the two-phase flow. Finally macro-scale simulations are performed for the whole absorption column using results from the previous scales as input data. The structured packing is treated as a porous domain to study liquid dispersion and the effect of geometrical features (i.e. walls and injectors). In this work we focus on the study of physical mass transfer at micro-scale.

A few papers dealing with CFD analysis of the physical and reactive mass transfer between a gas phase and a falling liquid film have been found in the literature. Haelssig et al. (2010) studied the effect of heat and mass transfer in multi-component ethanol–water liquid films using ANSYS® FLUENT for a 2D counter-current configuration. Haroun et al. (2010) have also investigated the effect of chemical reactions in 2D computational domains with an in-house VOF code called JADIM. Their results show a good match between the numerical predictions and the theoretical relationship between the Hatta number and the enhancement factor for both first and second order reactions. More recently, we developed a 3D VOF model to account for the physical mass transfer performance of structured packed columns at micro-scale for the oxygen–water system. The irregular liquid distribution over the metal plates (i.e. liquid misdistribution phenomenon) was linked with the actual oxygen mass transfer rate. The existence of an optimum working point as a trade-off between contact time and liquid spreading was predicted.

Haroun et al. (2014) applied a VOF numerical model to a meso-scale computational domain. The authors describe the gas–liquid flow within a set of actual Mellapak 250.X REUs. The values of liquid–solid contact angles were varied to mimic the geometrical imperfections of the plate surface. The results showed important variations of wetted area depending on the liquid–solid contact characteristics. Also the type of material that the structured packing is made of plays a vital role in the contact angle.

As a conclusion, there has been a substantial improvement in the CFD modeling of structured packed absorbers but experimental studies are still needed to verify numerical results. Fourati et al. (2012) carried out a visualization of the liquid misdistribution phenomena inside the absorber with gamma ray tomography. Recently Janzen et al. (2013) used X-ray tomography to study the influence of the liquid phase morphology on interfacial area and liquid hold-up. Kohrt et al. (2011) studied the influence of plate textures on physical mass transfer.

Our previous work focused on the validation of the hydrodynamics and physical mass transfer model. Also, the influence of several parameters such as liquid viscosity and plate textures on the hydrodynamics of the system was assessed. After the validation and the hydrodynamics study, the influence of the flow regime (i.e. trickling, rivulet and full film flow) on the mass

transfer performance was appraised. An optimum mass transfer point was observed when the transition to full film flow took place. Therefore, only the influence of the liquid velocity on the mass transfer performance was studied. Other parameters can be considered to be important from an industrial and a practical point of view though. The present paper expands our previous work in view of the effect of several process parameters on the gas absorption rate into falling liquid films. The parameters studied are liquid viscosity, gas velocity and pressure in both co- and counter-current configurations, contact angle, and plate surface textures. Further work will deal with the extrapolation of these results to higher scales in order to study the process parameters on actual REUs of structured packing material.

2. Model description

2.1. Hydrodynamics modeling

The volume of fluid (VOF) method is used to study the flow along an inclined plate that simulates the wall of a structured packing element. The VOF technique uses an additional transport equation to solve the volume fraction field of both phases. Volume fraction ranges from 0 to 1. Zero corresponds to a cell filled with gas phase and one corresponds to those filled with liquid phase. Computational cells whose volume fraction has values between 0 and 1 correspond to the interface, which is reconstructed using the Geo-Reconstruct algorithm.

The commercial software ANSYS® FLUENT v14.0 is used to carry out the simulations. The code uses the finite volume method to solve the Navier–Stokes equations (conservation of mass and momentum equations for incompressible and isothermal flow):

$$\nabla \cdot \vec{v} = 0 \quad (1)$$

$$\rho \left(\frac{\partial \vec{v}}{\partial t} + \vec{v} \cdot \nabla \vec{v} \right) = -\nabla P + \mu \nabla^2 \vec{v} + \rho \vec{g} + \vec{f} \quad (2)$$

The density and the viscosity in each cell are computed as volume fraction averaged:

$$\rho = F\rho_l + (1 - F)\rho_g \quad (3)$$

$$\mu = F\mu_l + (1 - F)\mu_g \quad (4)$$

The body force term in Eq. (2) considers only surface tension by the continuum surface force (CSF) theory (Brackbill et al., 1992) in the present model. CSF can lead to incorrect evaluations of the curvature term at the beginning of the simulations, giving abnormal predictions of the velocity at the interface. A variable time-step depending on a fixed value of the Courant number (i.e. which is set to be equal to 0.5) is used in this simulation to avoid this phenomenon (Min and Park, 2011). Surface tension by CSF model is calculated according to:

$$\vec{f} = \sigma \frac{\rho \kappa \nabla F}{0.5(\rho_l + \rho_g)} \quad (5)$$

2.2. Mass transfer modeling

Only molecular diffusion is considered as mass transfer mechanism. It is implemented as a source term (Eq. (8)) in the species transport equations (Eqs. (6) and (7)):

$$\frac{\partial(F_g \rho_g y_g)}{\partial t} + \nabla \cdot (F_g \rho_g \vec{v} y_g) + \nabla \cdot [D_g \nabla \cdot (F_g \rho_g y_g)] - S_{lg} = 0 \quad (6)$$

$$\frac{\partial(F_l \rho_l y_l)}{\partial t} + \nabla \cdot (F_l \rho_l \vec{v} y_l) + \nabla \cdot [D_l \nabla \cdot (F_l \rho_l y_l)] - S_{lg} = 0 \quad (7)$$

The source term is calculated according to Higbie's two film theory as

$$S_{lg} = k_l a_e C_l (y_l^* - y_l) = k_g a_e C_g (y_g - y_g^*) \quad (8)$$

The source term depends on the concentration gradient of a particular species between the bulk liquid phase and the interface, which is considered to be saturated (i.e. denoted by the superscript *). The solubility of a species is a function of the partial pressure and is calculated as

$$y_g^* = \frac{P_g}{He} \quad (9)$$

where He is called Henry's law constant.

The interfacial area is calculated according to the literature as the gradient of the volume fraction in a computational cell (Xu et al., 2009):

$$a_e = |\nabla F_l| = |\nabla F_g| \quad (10)$$

Higbie theory calculates the liquid side mass transfer coefficient as

$$k_l = 2 \sqrt{\frac{D_l}{\pi \tau}} \quad (11)$$

The exposure time is calculated using the following equation reported by Haroun et al. (2012) as

$$\tau = \frac{L_l}{|v_l|} \quad (12)$$

The interfacial velocity is obtained using Nusselt theory (Nusselt, 1916), which is valid for laminar flows. It calculates the velocity of the liquid as a function of the distance to the plate surface. The equation also considers the inclination angle of the plate with respect to the horizontal through the value of gravity:

$$v = \frac{\rho_l g \delta^2 \sin(\beta)}{2\mu_l} \left[1 - \left(\frac{z}{\delta} \right)^2 \right] \quad (13)$$

3. Results and discussion

3.1. Validation of hydrodynamics and mass transfer models

The validation of the present model for both hydrodynamics and mass transfer is discussed in our previous work (Sebastia-Saez et al., 2013). Hydrodynamics validation was carried out by evaluating the liquid film velocity profiles and the relationship between wetted area and the liquid injection velocity. The velocity profiles were compared to the Nusselt model prediction. The amount of wetted area was compared to the experimental data from Hoffmann et al. (2005), Iso and Chen (2011) and Iso et al. (2013). The general tendency by which the amount of wetted area increases with the liquid inlet velocity was also observed and the three regimes identified previously in literature (e.g. trickling, rivulet and full film flow) are well reproduced. The mass transfer model was validated by comparison with the experimental results from Xu et al. (2009), who studied the physical absorption of propane into a toluene liquid film in an inclined plate. The computational results show the correct tendency by which the amount of absorbed propane decreases with the liquid velocity due to the reduction of the exposure time. Further mass transfer validation consisted in the comparison with the theoretical models from Emmert and Pigford (1954), Zogg (1972) and Haroun et al. (2010), who presented correlations between the Sherwood and Reynolds numbers.

3.2. Effect of liquid viscosity

In this section the effect of the liquid phase viscosity on the absorption rate is assessed. The change in the liquid phase viscosity

Table 1
Viscosity values of amine solutions.

Amine concentration (%)	Dynamic viscosity (mPa s)
0 (pure water)	1
30	7.9
40	10.2

is achieved by varying the amine concentration in the aqueous solution.

The assumption of a mass weighted mixing law for the viscosity of the solution has been taken. The water viscosity is equal to 1 mPa s whereas the viscosity of pure MEA has been taken from data provided by Dow® Chemical Company at 295 K (i.e. 24 mPa s). Table 1 shows the values of viscosity used for the solutions in the simulations. The values have been compared to the work of Amundsen et al. (2009) who presented data about the density and viscosity of aqueous MEA solutions. The authors conclude that the viscosity of the solution depends on the amount of CO₂ dissolved. The assumption of neglecting the variation in viscosity and density due to the amount of dissolved gas has been taken in this work. It has also been observed that the value of viscosity used for pure MEA in Amundsen et al. (2009) is lower than the one used in this work.

Viscosity can affect the mass transfer source term through both solubility and the diffusion coefficient. The influence on the solubility can be neglected according to Wang et al. (2013). The diffusivity coefficient of oxygen in a MEA aqueous solution can be obtained using the Wilke–Chang equation (Wilke and Chang, 1955):

$$D = \frac{7.4 \times 10^{-8} (\phi M)^{1/2} T}{\mu V^{0.6}} \quad (14)$$

where the association parameter of water (ϕ) is equal to 2.6.

Fig. 1 shows the effect of the amine concentration on the oxygen absorption rate for two different liquid loads. Two solutions commonly found in industry i.e., 30% and 40% by weight aqueous MEA are evaluated. The liquid injection velocities considered are 38 cm³ cm⁻² s⁻¹ and 45 cm³ cm⁻² s⁻¹, which correspond to the rivulet flow regime (Sebastia-Saez et al., 2013; Hoffmann et al., 2005). Rivulet flow is a commonly found regime in industrial absorbers. As a general trend the oxygen absorption rate decreases as MEA concentration increases. This can be explained by the effect of viscosity on the gas diffusivity. According to Eq. (14) the diffusion coefficient decreases as viscosity values increase. The same trend is obtained when the liquid load increases. The rate of absorption is improved when the liquid load goes from 38 to 45 cm³ cm⁻² s⁻¹. This occurs because of the higher liquid spreading related to higher

liquid injection rates in the rivulet flow regime. The graph shows that the effect of the liquid load on the absorption rate is small compared to the effect of viscosity for the range of values considered. A similar result was also found by Janzen et al. (2013).

The effect of viscosity on the absorption rate can also be explained in terms of the relationship between wetted area and the ratio of cohesive and distortive forces. An inverse relationship exists between viscosity and surface tension (Singh and Singh, 2011). Therefore an increase in viscosity causes a decrease in surface tension, which gives a bigger Weber number. The increase in Weber number leads to an improved spreading of the liquid over the plate. However, the effect of liquid injection velocity has been proven to have less impact than the liquid viscosity for the range of values considered.

Fig. 2 shows the transient behavior of the oxygen absorption process. The graph confirms the tendency by which a higher MEA concentration gives a lesser amount of oxygen absorbed. A difference in the averaged oxygen mass fraction for both liquid loads is found for the case of pure water whereas this does not happen for both aqueous MEA solutions. Therefore, no substantial difference is found between the liquid injection velocity cases with the same value of the liquid viscosity for the aqueous MEA solutions. This can be explained by the fact that a lower value of diffusivity requires more time for diffusion to take place. Also the liquid injection rate is high enough to make the liquid film development faster than diffusion, which also contributes to this behavior.

The transient graph shows three different regions for the case of pure water. This has already been reported in literature (Sebastia-Saez et al., 2013). The first region is characterized by a linear relationship between time and oxygen absorption rate. It is caused by the fact that a droplet is formed in the surroundings of the liquid inlet and it begins to fall down the plate as soon as its weight overcomes surface tension. When this happens distortive forces are greater than cohesive forces and the spreading of the liquid is initiated. This marks the beginning of the liquid film formation. The first stage takes generally a longer time in the case of the amine solutions. This can be explained because a higher viscosity reduces the Reynolds number, which in turn is a measure of the quantity of inertia of the liquid. Hence a lower inertia of the liquid results in more time for the liquid to commence falling along the plate.

The duration of the second stage is inversely proportional to the liquid injection velocity. The analysis has been carried out for injection velocity values that correspond to the rivulet flow regime. As a consequence, a higher inertia in the fluid results in better spreading of the liquid. Higher averaged oxygen concentration at pseudo-steady state along with less time needed to reach full film development results in higher oxygen absorption rates.

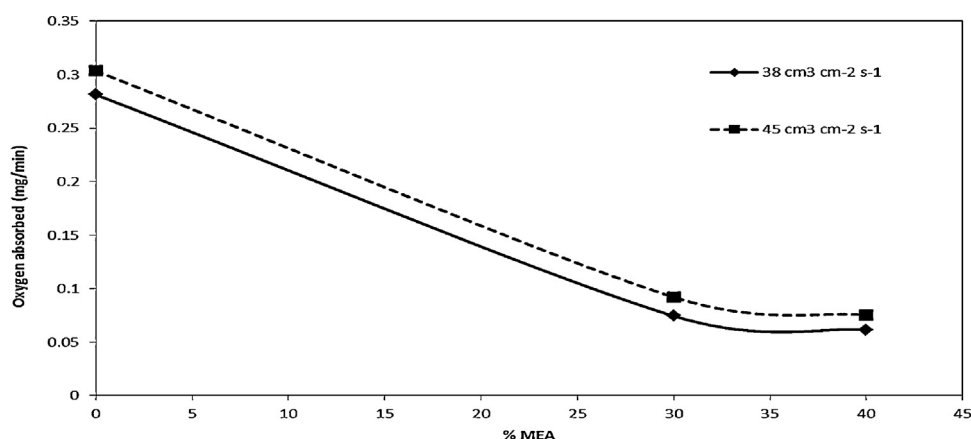


Fig. 1. Effect of MEA solution on the oxygen absorption rate for various liquid loads.

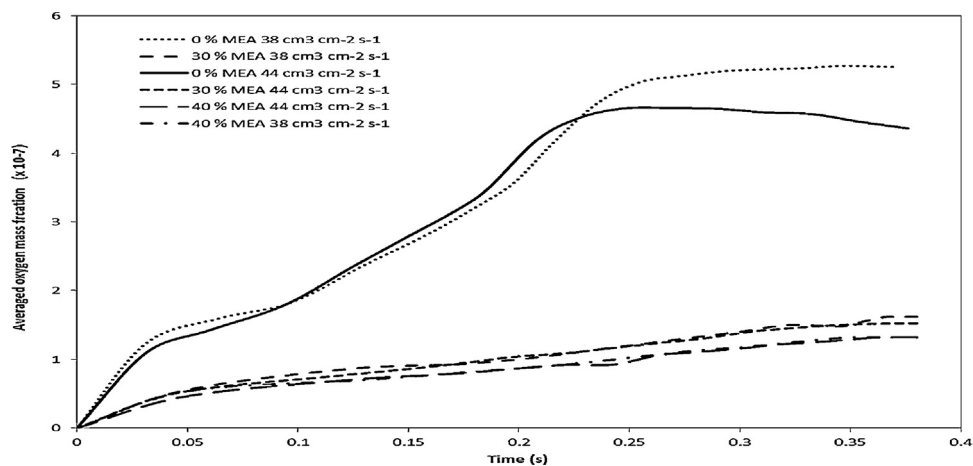


Fig. 2. Averaged absorbed oxygen mass fraction vs. time for different MEA concentrations with different liquid loads.

However the absorption rate in the second region is slower than in the first one. This is caused by a smaller mass fraction of oxygen within the bulk of the liquid film compared to the first region. Finally the pseudo-steady state is achieved, which constitutes the third region. This stage is characterized by a constant oxygen absorption rate, mainly dependent upon the amount of wetted area.

The boundaries between the aforementioned three regions are not so clear for the case of the amine solutions. The transitional behavior can be divided in two regions. The second region is smoother and slowly approaches the pseudo-steady state. This is caused by the presence of MEA, which gives a less chaotic hydrodynamic development of the liquid film. Also the variation of the absorption rate is slower for the aqueous solutions due to the decrease in diffusivity associated with high viscosity.

It can be concluded that an increase in the liquid phase viscosity implies a reduction in the oxygen absorption rate. However, an increase in the amine concentration carries also a higher probability for reactive mass transfer to occur. The presence of the chemical reaction enhances the absorption according to the Hatta number (Haroun et al., 2010). Therefore two opposite effects are found depending on the amine concentration, which could lead to the existence of an optimum point. This feature needs to be investigated in future studies.

3.3. Effect of gas phase pressure

The influence of the gas pressure on the oxygen absorption rate is investigated in this section. Pressure affects mass transfer via its effect on solubility. The relationship between both parameters is implemented in this study by Henry's law. The three values of pressure tested are 101,325 Pa (i.e. 0 bar), 151,987 Pa (i.e. 0.5 bar) and 202,650 Pa (i.e. 1 bar) gauge pressure. Their corresponding solubility values are shown in Table 2 and are incorporated to the UDF code. The values of solubility are calculated using Henry's law at a constant temperature of 303 K. A pressure increase causes a bigger difference between the interfacial and bulk concentration of the gas phase resulting in bigger values of the source term.

Table 2
Solubility of oxygen in water used in the present study.

Relative pressure (bar – Pa)	Solubility in mass fraction (Atkins, 1998)
0.0 – 101,325	8.5 10 ⁻⁶
0.5 – 151,987	13.5 10 ⁻⁶
1.0 – 202,650	17.0 10 ⁻⁶

Fig. 3 depicts the influence of pressure on the transient behavior of the mass transfer rate. The simulations are carried out for pure water at a liquid load of 38 cm³ cm⁻² s⁻¹ with oxygen as stagnant gas phase. As a general trend a higher pressure gives a higher amount of oxygen absorbed during all the stages of the liquid film development.

The absorbers used for post-combustion carbon capture work at atmospheric pressure though. The use of pressurized scrubbers could be considered to be applied since an increment of 0.5 bar in the gas pressure gives an improvement of up to 60% in the amount of oxygen absorbed.

However, the pressurization of the reactor could be seen as a drawback due to maintenance and safety reasons.

3.4. Effect of gas phase inlet configurations and its velocity

In this section the effect of the gas velocity on the transient behavior of the oxygen absorption is investigated. The gas velocity can be defined by the f -factor (Armstrong et al., 2013; Said et al., 2011), which is calculated as

$$f = v\sqrt{\rho} \quad (15)$$

Velocity profiles are specified at the gas inlet with user-defined functions (UDFs) considering continuity of the tangential velocity component and zero shear stress at the interface (Zakaria and Gamiel, 2012; Kabova et al., 2014). Atmospheric pressure and an f -factor equal to 4 are used in both co-current and counter-current configurations. For the counter-current calculations the same velocity profiles with negative values are used (Fourati et al., 2013).

The results show that the mass transfer is increased if momentum is applied to the gas phase. A certain amount of drag exists between the gas and the liquid phase, which affects the liquid film hydrodynamics. This results in higher and lower values of the wetted area for the co- and counter-current configurations respectively. The co-current configuration flattens the liquid film helping to its development whereas counter-current tends to hold the liquid near the inlet hindering its development. Flow configurations also affect the contact time between phases. Also co-current flow augments contact time, which helps to improve the absorption. Fig. 4 shows the comparison between the transient evolution of the amount of oxygen absorbed for stagnant, co- and counter-current flows. It is observed that both co- and counter-current configurations give an enhancement in the absorption rate with respect to the stagnant case. The gain is more pronounced for the co-current disposition.

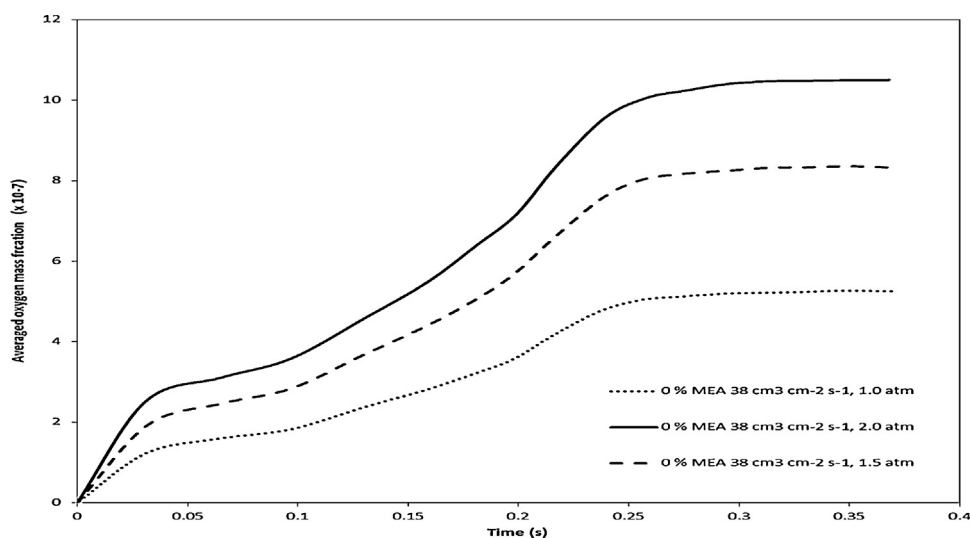


Fig. 3. Effect of gas phase pressure on transient behavior of averaged oxygen mass fraction.

The transient behavior shows three different regions for all cases as it was already discussed in previous sections. The transition between regions is smoother for the co-current case because the gas phase drags the liquid forcing it to fall down the plate hence helping to the development of the liquid film. In this sense, drag force would also act as a distortive force in the same way as inertia. The liquid rivulets are flattened and the fully developed liquid film covers more area than in the counter-current and stagnant cases. The initial accumulation of liquid near the inlet is downsized hence reducing its effect on the mass transfer process.

Fig. 5 shows the combined effect of gas pressure and velocity. It is shown that an increment on the absorption rate is found when both variables increase. The figure shows the absolute rate of absorption in mass per unit time at pseudo-steady state. All data series run in parallel and equidistant to each other, suggesting a linear relationship between the absorption rate and the flow configuration. Also, the relationship between the absorption rate and the gas pressure presents a linear behavior.

As a conclusion the importance of gas velocity and flow disposition on the overall process has been highlighted. Co-current flow is proven to be more beneficial than the counter-current configuration for inclined metallic plates. Co-current configuration causes

a higher liquid spreading due to the interfacial drag, which affects the contact area between phases. However counter-current flow is the most common configuration found in industry for absorption columns. Similar studies on the behavior of both flow configurations considering a set of REUs are needed to check the convenience of counter-current flow as the preferred choice.

3.5. Effect of liquid–solid contact angle

This section shows how contact angles affect the wetting behavior. Contact angles depend on the interaction between the solid and liquid molecules. High liquid–solid affinity results in good adhesion hence contact angles below 90° . The opposite takes place when intermolecular forces within the liquid phase are greater than the solid–liquid interaction. The analysis of the relationship between wetted area and the Weber number only considers surface tension (i.e. liquid–liquid interaction or cohesion) but neglects adhesion forces. It is reported in literature that flow patterns evolve from rivulet to full film flow when the contact angle is reduced from 70° to 10° (Haroun et al., 2014). Strong adhesion can be considered to be a distortive force as inertia or gravity, which tends to increase the

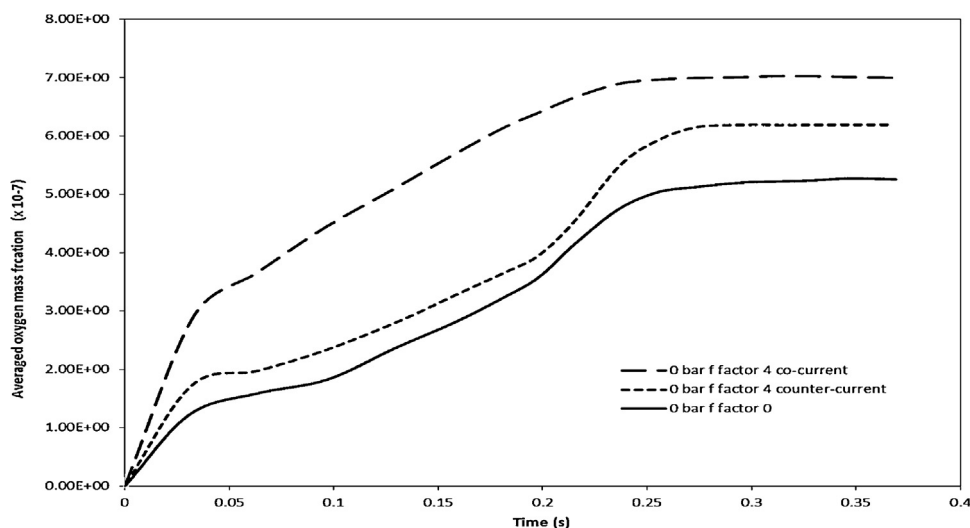


Fig. 4. Effect of gas phase velocity configurations on transient averaged oxygen mass fraction at atmospheric pressure.

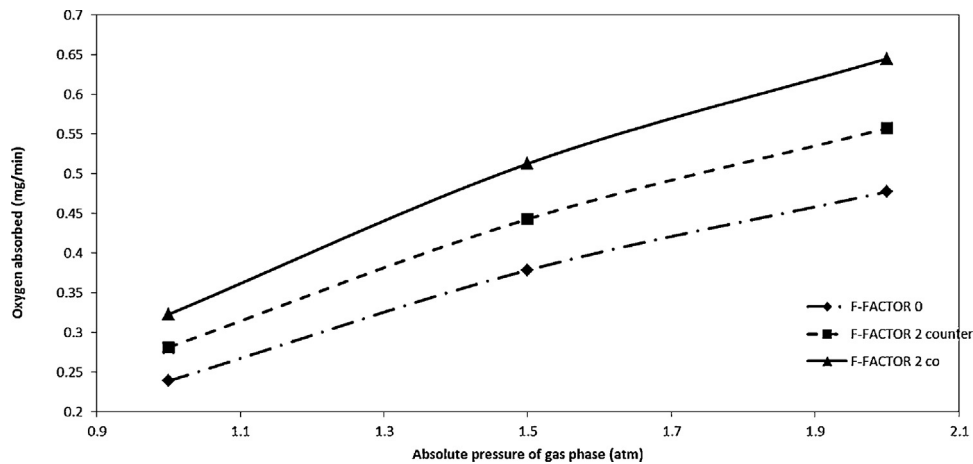


Fig. 5. Effect of gas phase pressure on oxygen absorbed for different f -factors.

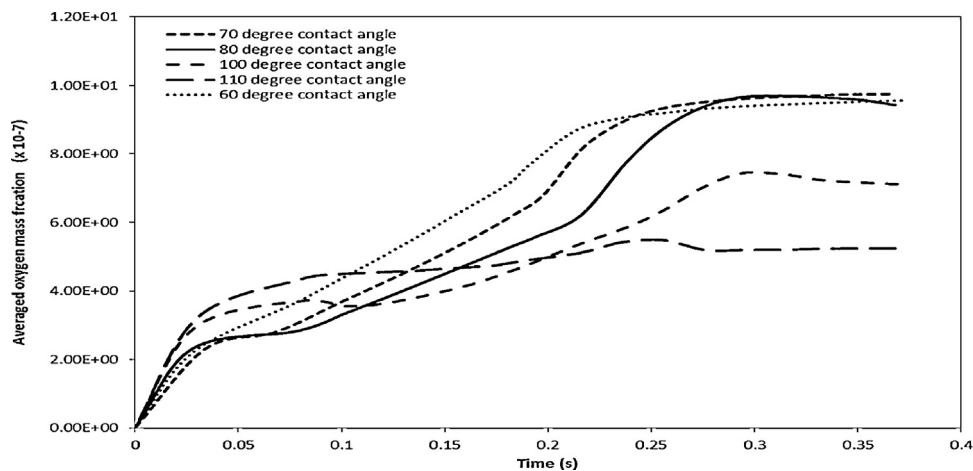


Fig. 6. shows the averaged oxygen mass fraction at outlet conditions for several wetting angles.

interfacial area, whereas poor adhesion reinforces cohesive effects. Weber number is then incomplete to explain liquid spreading since it only considers surface tension as a cohesive force.

The contact angles tested in this study are 60, 70, 80, 100 and 110° to assess wetting and non-wetting conditions. The simulations are performed for a liquid load of $38 \text{ cm}^3 \text{ cm}^{-2} \text{ s}^{-1}$ (i.e. rivulet flow regime), a relative pressure of 0.5 bar (151,987 Pa), and a gas phase in counter-current flow (f -factor 2). Fig. 6 shows the transient behavior of the averaged oxygen concentration within the liquid film. Each data series corresponds to a particular value of the contact angle. There is no substantial variation in the mass transfer rate at pseudo-steady conditions for wetting angles ($\theta < 90^\circ$). However there is a decrease in the oxygen concentration caused by the reduction in wetted area for non-wetting contact angles ($\theta > 90^\circ$). Fig. 7 shows that only the central channel appears with no lateral rivulets for contact angles greater than 90°.

A permanent formation and detaching of the central rivulet occurs similarly to the trickling flow for a contact angle of 110°. The trickling flow is characterized by the prevalence of cohesive over distortive forces (i.e. low Weber number). A poor liquid spreading is encountered for non-wetting conditions despite the fact that the liquid injection velocity is high enough to achieve full film flow regime.

It can be concluded that a huge decrease in the oxygen transfer rate is found when the contact angle is increased over 90° due to

a fundamental wetting behavior. Hence it is important to ensure a good wetting condition through a proper selection of the packing material.

3.6. Effect of plate textures

The relationship between the geometrical pattern on the plate surface and the gas–liquid interfacial area was assessed in our previous paper (Sebastia-Saez et al., 2013). We concluded in that study that a proper surface texture in the metallic plate can influence

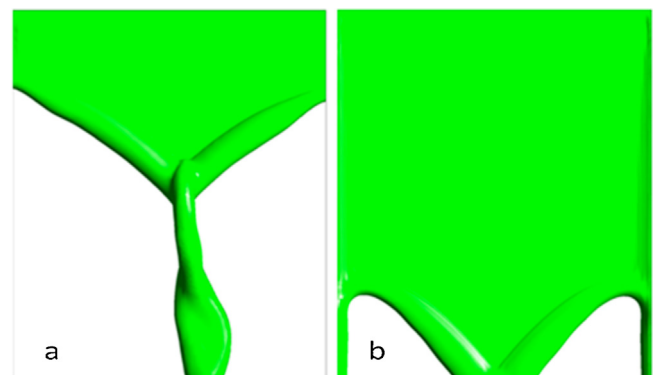


Fig. 7. Film flow patterns for 100° (a) and 70° (b) contact angles.

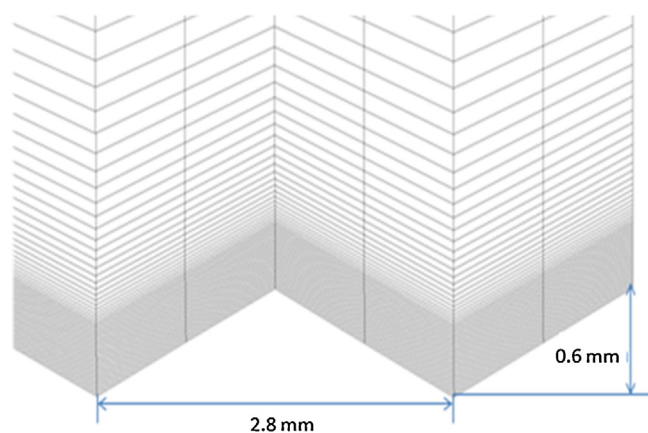


Fig. 8. Detail of the mesh and the triangular pattern used as surface texture.

Table 3
Oxygen absorption rate as a function of a particular geometrical pattern.

Geometrical pattern	Oxygen absorption rate (mg/min)	
	$25 \text{ cm}^3 \text{ cm}^{-2} \text{ s}^{-1}$	$38 \text{ cm}^3 \text{ cm}^{-2} \text{ s}^{-1}$
Streamwise	0.154	0.246
Smooth surface	0.152	0.239
Perpendicular	0.149	0.233

the amount of wetted area hence the mass transfer performance. Therefore, in this work the study is expanded to investigate the effect on the oxygen absorption rate. Two types of geometrical patterns, along with the smooth plate case were tested. The textures consisted of a triangular wavy pattern forming channels in both a stream-wise and perpendicular to the flow disposition. Further detail about the texture patterns can be found in Fig. 8. The triangle has a 2.8 mm base and 0.6 mm height. The computed data can be found in Table 3.

An improvement in mass transfer rate is achieved for both liquid injection rates tested (38 and $25 \text{ cm}^3 \text{ cm}^{-2} \text{ s}^{-1}$) when the channels are disposed in the same direction as the liquid flow. The canalization of the fluid along the plate tends to avoid the effect of cohesive forces. A perpendicular disposition constitutes a barrier for the hydrodynamic development of the liquid film allowing cohesive forces to act and increasing the time to achieve pseudo-steady state. This behavior is in accordance with the results observed in the authors' previous paper for the percentage of the plate area covered by the liquid. The stream-wise pattern is the best option to increase the gas–liquid interfacial area whereas the perpendicular disposition diminishes it. However, the oxygen absorption rate has been found to be small (i.e. with differences not higher than 3%) in the case of an inclined plate of this size. As a conclusion it can be said that the selection of the plate texture can influence the liquid spreading and the mass transfer rate. However, further work should be carried out in bigger computational domains to assess this effect in actual REUs.

4. Conclusions

In this work the influence of several parameters on the oxygen mass transfer into a liquid film is assessed through numerical modeling. The parameters are liquid phase viscosity, gas pressure, gas velocity, co- and counter-current flow configurations, liquid–solid contact angle and plate textures.

The influence of the liquid phase viscosity has been tested by varying the concentration of monoethanolamine in the aqueous solution. Higher amine concentration leads to higher viscosity which diminishes the efficiency of mass transfer.

Gas pressure, gas velocity and flow configurations (i.e. co- and counter-current) are also proven as key factors for the mass transfer performance. Gas pressure directly affects the value of solubility, which results in an absorption enhancement. Pressurization is then proven to be beneficial for the process since a gain in the oxygen absorption rates of up to 60% is found. A gas phase with a specified inlet velocity in both co- and counter-current configurations also improves the mass transfer process compared to the stagnant case. The interfacial drag maximizes the liquid spreading for the co-current case giving more exchange area available for mass transfer.

The effect of contact angles between the liquid phase and the solid surface has also been evaluated. The importance of a good wetting condition is discussed. Contact angles over 90° reduce the amount of wetted area, eliminating lateral rivulets and increasing the height of the central one. The study highlights the importance of the liquid–solid interaction as a distortive force that improves the amount of interfacial area.

Finally the influence of the plate texture is tested, confirming the fact that it can either enhance or hinder the process through its effect on the wetted area.

CFD has been proven to be a suitable tool to analyze the flow characteristics between metal sheets within a structured packing. However more numerical and experimental studies are needed to gain a thorough understanding of the absorbing process that would lead to better designs.

Acknowledgments

The authors gratefully acknowledge the financial support for this work by the UK Engineering and Physical Sciences Research Council (EPSRC) project grant: EP/J020184/1 and FP7 Marie Curie iComFluid project grant: 312261.

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2014-09-01T00:00:00Z

Elsevier

<http://dx.doi.org/10.1016/j.ijggc.2014.06.029>

Sebastia-Saez D, Gu S, Ranganathan P, Papadikis K. (2014) Micro-scale CFD study about the influence of operative parameters on physical mass transfer within structured packing elements. International Journal of Greenhouse Gas Control. Volume 28, September 2014, pp. 180-188